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FINITE ELEMENT PREDICTION OF THREE-DIMENSIONAL CARBON DIOXIDE DIFFUSION IN GRAIN BINS

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ABSTRACT

A three-dimensional diffusion equation in the Cartesian coordinate system was solved using the finite element method for predicting CO_2 distribution in grain bins. The predicted CO_2 concentrations were compared with experimental data on the movement of CO_2 through wheat contained in 1.45-m-diameter model bins. Variables considered in the experiments included three different floor openings (circular near the centre, circular near the wall, and rectangular), open and covered grain surfaces, and different CO_2 concentrations at the perforated openings. The predicted CO_2 concentrations were much lower than the measured data for simulations starting from time 0. The underpredictions were observed because of the bulk flow of CO_2 through wheat when the dry ice sublimated into CO_2 gas. The accuracy of predictions improved when the simulations were run using the measured data at 1 h as the initial condition.

INTRODUCTION

Insects, mites and fungi can cause considerable damage to the grain stored in storage units of various shapes and sizes. Stored grain can be protected from insects and mites using chemicals such as contact insecticides and acaricides, and can be disinfested by fumigation. These chemicals leave objectionable chemical residues on the grain and are hazardous to handle and apply. Also, many stored-product pests are developing resistance to chemicals.

Due to perceived carcinogenicity to mammals, ethylene dibromide and other liquid fumigants were banned by the Canadian federal regulatory agency in 1984, followed by an on-going regulatory review of remaining chemicals such as phosphine and methyl bromide. Therefore, alternative ways of protecting stored grain against pests should be explored.

Controlled atmosphere (CA) storage is a potential alternative method of insect control. In principle, CA storage is an artificially created storage where the gaseous composition of the intergranular air is altered by injecting either CO_2 or nitrogen (N_2) to create an atmosphere lethal to pests (Banks and Annis, 1977). CO_2 is more effective than N_2 in controlling insects (Jay, 1980). Effectiveness of CAs for the control of various stored-product pests depends on several factors, including temperature and moisture content of the grain, gaseous composition, species and life stage of the pests, and exposure time.

For successful control of pests using CAs, gases should be introduced into grain at desirable moisture contents and temperatures. The gases should be distributed uniformly in the grain bulk to maintain adequate concentrations for the required exposure time in all locations of the bin. Uniform distribution of the introduced gases is dependent on the rate of movement of these gases through the grain bulk, which, in the absence of pressure and temperature gradients, depends on the rate of diffusion (Cunningham and Williams, 1980). An understanding of the distribution of gases in a stored grain bulk is needed for engineering design and successful application of CA storage to control pests. Limited research has been done on modelling the diffusion of CO_2 in masses of grain (Singh *et al.*, 1983; Jayas *et al.*, 1988). Both of these models are axisymmetric models. In most farm bins, CO_2 produced by the metabolism of grain and pests at various locations in the bulk may be sorbed by the grain and lost through bin structure at many locations. Also, CO_2 may be introduced for pest control at points near the wall, making CO_2 diffusion non-axisymmetric. Also, an axisymmetric model cannot predict the movement of CO_2 in non-circular bins. Only a three-dimensional model can predict potential CO_2 distribution in the grain bulk under these circumstances.

The objective of this study was to model the three-dimensional diffusion of CO_2 in stored grain bulks using the finite element method, and to compare simulated results with experimental data on the movement of CO_2 in model bins containing wheat.

MODEL DEVELOPMENT

The partial differential equation governing the unsteady state diffusion of gases in an anisotropic porous media in the Cartesian coordinate system is given by (Geankoplis, 1983) (list of symbols at end of paper):

$$\frac{\partial}{\partial x}\left(D_x \frac{\partial c}{\partial x}\right) + \frac{\partial}{\partial y}\left(D_y \frac{\partial c}{\partial y}\right) + \frac{\partial}{\partial z}\left(D_z \frac{\partial c}{\partial z}\right) + q = \frac{\partial c}{\partial \tau} \qquad [1]$$

subject to the boundary conditions,

$c = c_{S_1}$ on S_1 [2]

$c = c_{S_2}$ on S_2 [3]

$$D_x \frac{\partial c}{\partial x} \ell_x + D_y \frac{\partial c}{\partial y} \ell_y + D_z \frac{\partial c}{\partial z} \ell_z + Q = 0 \qquad \text{on } S_3 \qquad [4]$$

and the initial condition, $c(x,y,z,\tau=0) = c_i$ on Ω [5]

S_1, S_2 and S_3 together make the total boundary of the domain W. For a grain bin, S_1 is the portion of the boundary through which CO_2 is injected into the bulk, S_2 is the portion of the boundary through which the intergranular air leaves the grain bulk and S_3 is the bin wall.

METHODS AND MATERIALS

Test bins

To validate the CO_2 diffusion model, CO_2 distribution tests were conducted in model bins filled with wheat. Three 1.45-m-diameter and 1.47-m-high bins were obtained from a local bin manufacturer (Westeel, Winnipeg, Manitoba). Three different, partially-perforated floor openings were installed in the bins. The floor openings were a 0.3-m-diameter opening at the centre of bin 1, a 0.3-m-diameter opening near the wall of bin 2, and a 1.22 m x 0.46 m rectangular opening in bin 3. Metal boxes having volumes of 0.09 m^3 for bins 1 and 2 and 0.2 m^3 for bin 3 were fabricated and mounted centrally under the perforated floor openings of the bins. A known quantity of dry ice, the solid form of CO_2, which sublimates into CO_2 gas at temperatures above -78.7°C was placed in the boxes to render CO_2 gas. The dry ice introduction boxes were equipped with a 7.5-cm-diameter PVC pipe fitting and screw cap for placing the dry ice and for aerating the grain after each replication. The bins were placed on 50-cm-high wooden platforms. All the joints and bolt holes in the bins were made gas-tight using silicon sealant.

Gas samples were drawn from five levels, spaced 0.33 m apart in a vertical direction, using 3.2-mm O.D. nylon pressure tubing fitted with rubber septa at the outer end. There were 11, 12, and 13 sampling points at each level for bins 1, 2, and 3, respectively (Fig. 1). Gas samples, collected using 10-mL syringes, were analyzed for CO_2 concentrations using a Perkin-Elmer Sigma 3B gas chromatograph equipped with a thermal conductivity detector and a 1 mL fixed volume injection loop. The grain temperatures at 5 locations in bin 1 and 15 locations in each of bins 2 and 3 were recorded using copper-constantan thermocouples and a temperature indicator. In addition, temperatures were also measured at 2 locations in the boxes.

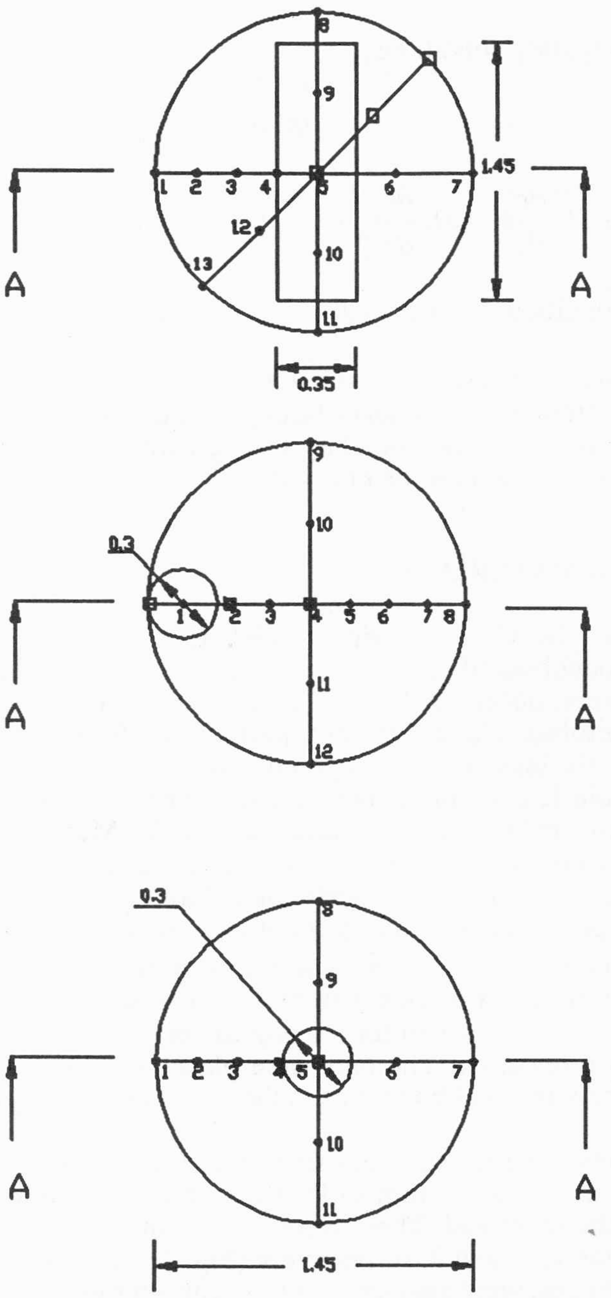

Fig. 1: Temperature (❑) and CO_2 (●) sampling locations. Top: rectangular opening, Centre: 0.3-m-diameter opening near the wall, bottom 0.3-m-diameter opening near the centre (all dimensions in m).

Experimental procedure

The bins were filled with Canadian red spring wheat (cv. "Katepwa") graded No. 1 by the Canadian Grain Commission. The wheat had a dockage content of 0.5% and moisture content of 12.8%. The bins were filled to a depth of 1.37 m by manually pouring the wheat from buckets. For bins 1 and 2, 180 g of dry ice, that would create a CO_2 concentration of approximately 10% in the intergranular air space of the wheat bulk was placed in the box and for bin 3, 370 g of dry ice, that would create a CO_2 concentration of approximately 20% was used. Samples of intergranular air were collected and analyzed for CO_2 concentration at 1, 3, 6, 12, and 21 h, after the introduction of the dry ice. In all the bins, three replicates with an open grain surface and three replicates with the grain surface covered with a 0.2-mm-thick plastic sheet were made. After each replicate, the grain was aerated using a 1.5 kW centrifugal blower (General Blower Co., Wheeling, Illinois, USA), for approximately one hour to bring the CO_2 concentrations in the grain bulk to atmospheric level (about 0.03%), and left undisturbed for about 24 h before the start of the next experiment. The blower was run for another 15 min just before the start of the next experiment.

Simulation Procedure

A FORTRAN program was written to solve the unsteady state diffusion problem (Eq. 1). The program can handle linear and quadratic hexahedron elements with 1, 2 or 3 point Gauss quadrature integration in each plane. The boundary conditions may include specified CO_2 concentration on the portions of the boundary ($c=c_s$), and no flow ($\partial c/\partial n = 0$) or specified flow ($\partial c/\partial n =$ constant) across the boundary.

For simulating the distribution of CO_2 in the grain bulk, half of the grain bulk (along section AA of Fig. 1) was discretized into linear elements with 445 nodes for bin 1, 430 nodes for bin 2, and 390 nodes for bin 3. The measured CO_2 concentrations near the floor opening (sampling point 5 of bin 1, 1 of bin 2, and the average of sampling locations 5, 9, and 10 of bin 3) were specified for the nodes lying inside or on the boundary of the floor opening. To include this boundary condition in the program, the measured CO_2 concentrations at these locations were fitted to an equation of the form (SAS, 1982):

$$CC = A\, e^{-B.t} \qquad [6]$$

For simulation of the tests with uncovered top grain surfaces, the measured CO_2 concentration near the surface of the grain was specified at the nodes lying on this boundary, and when the grain surface was sealed with a plastic sheet, this boundary was assumed to be impermeable to CO_2 diffusion ($\partial c/\partial n = 0$). The bin wall and the bin floor, excluding the floor opening were assumed to be impermeable to the flow of CO_2. A diffusion coefficient of 4.15 mm^2/s for red spring wheat at 12% moisture content (m.c.) (Singh *et* *al.*, 1985) was used in the simulations. It was assumed that the diffusion coefficient was independent of the direction of diffusion (Singh *et al.*, 1984), and of concentration (Cunningham and Williams, 1980). As the wheat used in the experiments was dry (m.c. less than 14.5%), the CO_2 adsorption and production by wheat was assumed to be negligible (q=0) (White *et al.*, 1982).

RESULTS AND DISCUSSION

Experimental data

Fig. 2 shows the contour plots of measured CO_2 concentrations at 12 h (mean of three replicates) along section AA of bins 1, 2, and 3. The CO_2 concentrations became nearly uniform in the top two-thirds of the grain bulk. This indicates that even if CO_2 is introduced at a corner of the grain bulk (as in bin 2), the concentrations will become uniform in upper layers of the bulk. Thus, in existing bins with a non-perforated floor, a hole could be made near the bin wall to introduce CO_2. This conclusion cannot be extrapolated for large bins without further experimental studies.

The objective of CA treatment of stored grain is to create high CO_2 concentrations at all points in the grain bulk for effective control of insects. When dry ice was introduced through floor openings, high CO_2 concentrations were usually observed near the floor openings. As our findings indicate, the CO_2 concentrations are lower at points away from the floor opening in the lower portions of the grain bulk and at all points in the upper half of the bulk. When the grain surface was not covered, the CO_2 concentrations in the top layers were much lower than when the grain surface was covered with a plastic sheet. For example, in bin 1, the 2% CO_2 concentration line was approximately 1/3 of its height below the surface of the grain when the grain surface was uncovered, as compared with its level near the grain surface when the grain surface was covered with a plastic sheet. Thus, in a CA treatment it is advantageous to cover the grain surface to achieve high CO_2 concentrations in the top layers of the grain bulk.

Simulation results

The simulations were run with the assigned boundary conditions and the initial concentration at every node in the grid set equal to the atmospheric CO_2 concentration (0.55×10^{-3} kg/m^3). The predicted CO_2 concentrations were much lower than the measured concentrations at every sampling point and at all times. The mean relative percent error of prediction was calculated as:

$$e = \frac{1}{n}\sum\frac{|M_i - P_i|}{M_i} \times 100 \qquad [7]$$

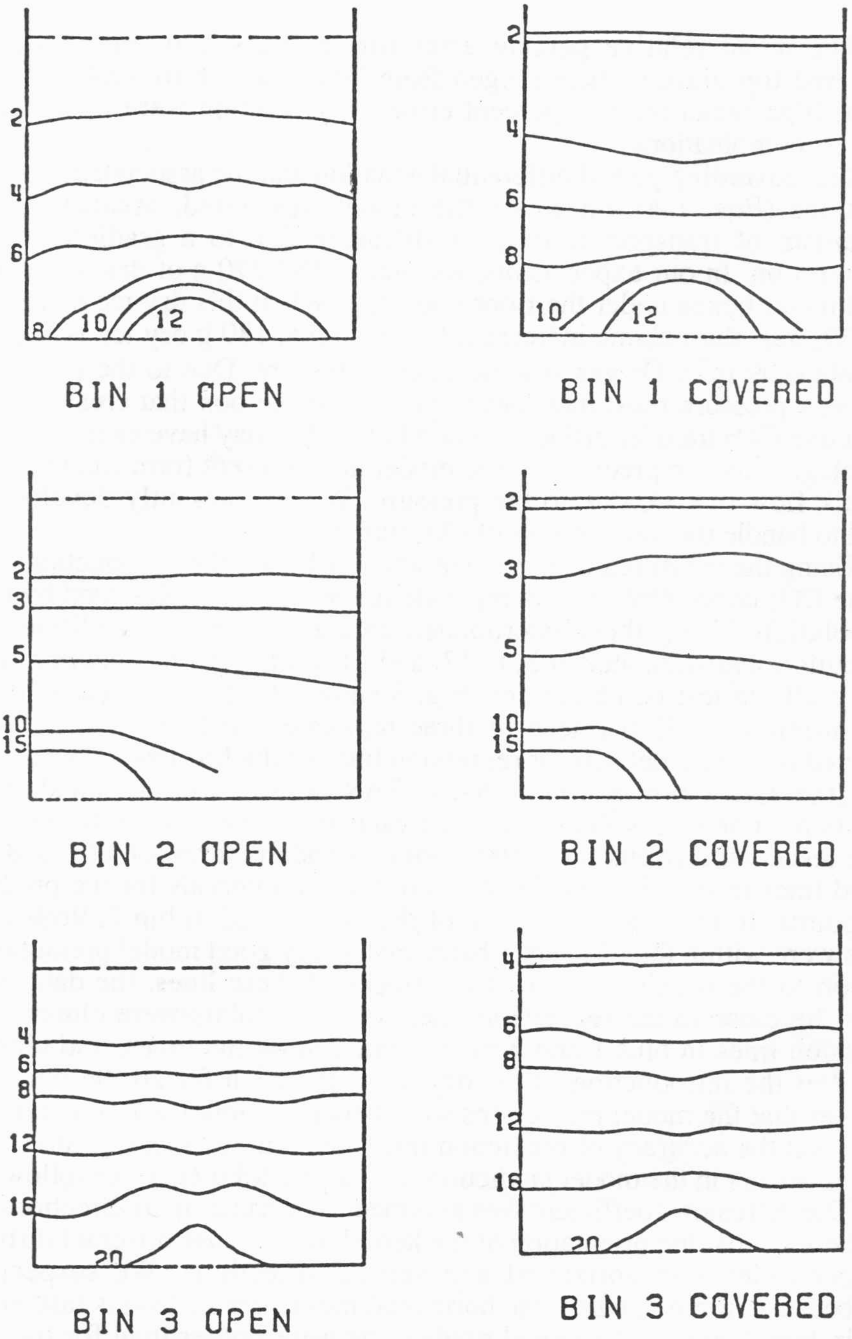

Fig. 2: Iso-concentration lines of CO_2 (%) at 12 h (drawn using the mean of measured concentrations from three replicates), along section AA of bins 1, 2, and 3. The dashed lines at the bottom show the floor opening and the horizontal lines at the top show the grain surface.

The mean relative percent error for replicate 1 of bin 1 with an uncovered top grain surface ranged from 70.8% at 3 h to 30.9% at 21 h. Similar high mean relative percent errors in prediction were observed for other test combinations.

The governing partial differential equation and the associated boundary conditions (Eqs. 1-4), on which the model was based, assume that the mechanism of transport is only by diffusion due to a gradient in CO_2 concentration. In our experiments, we placed 180-370 g of dry ice in well-sealed metal boxes under the floor openings. When this dry ice sublimated into CO_2 gas, the volume increased. For example, 180 g dry ice is expected to create 0.09 m^3 CO_2 gas at atmospheric pressure. Due to the increase in volume, a pressure must have developed inside the box that created forced convective CO_2 transfer inside the grain bulk. This may have caused the high percentage errors in prediction. The model in its present form cannot handle the bulk flow of CO_2 due to the pressure. We are presently developing a model to handle the initial stages of CO_2 introduction.

Using the measured CO_2 concentrations 1 h after the introduction of dry ice, the CO_2 concentration at every node in the grid was calculated by linear interpolation. Using these interpolated data as the initial condition, CO_2 distribution was simulated at 3, 6, 12, and 21 h after the introduction of dry ice, for all the test combinations. Fig. 3 shows the plot of measured CO_2 concentrations at 21 h (mean of three replicates) and the concentrations predicted by the model. Linear regression lines of the form y=mx were fitted using the GLM procedure of SAS (SAS, 1982). For an ideal model prediction, these slopes should have the value of 1. The mean of the slopes of all the test combinations was 0.9869 with a standard deviation of 0.048. The dashed lines in Fig. 3 show the 95% confidence intervals for the predicted data points. In bin 1 and 3, 97.5% of the points, and in bin 2, 96% of the points were within the 95% error band. For a very good model prediction, in addition to the proximity to 1 of the slopes of these lines, the data points should lie close to the regression line. The data points were closer to the regression lines in bins 1 and 3 than in bin 2 at all the times, and closer at 21 h after the introduction of the dry ice than at 3 h for all the bins. This indicates that the model predictions were better for bins 1 and 3 than for bin 2, and that the accuracy of prediction improved with increase in time for all the bins. Errors in the model predictions could be attributed to the following:

1. The diffusion coefficient was assumed to be same in all directions. We believe that the orientation of the kernel might cause different diffusion coefficients in horizontal and vertical directions. We suspect this because in bin 2, where the horizontal movement of CO_2 is farther than in bins 1 and 3, the model predictions were poorer than for the other two bins. In all the bins at points away from the floor opening near the lower portions of the grain bulk, the predicted CO_2 concentrations were lower than the measured concentrations. Furthermore, it has been

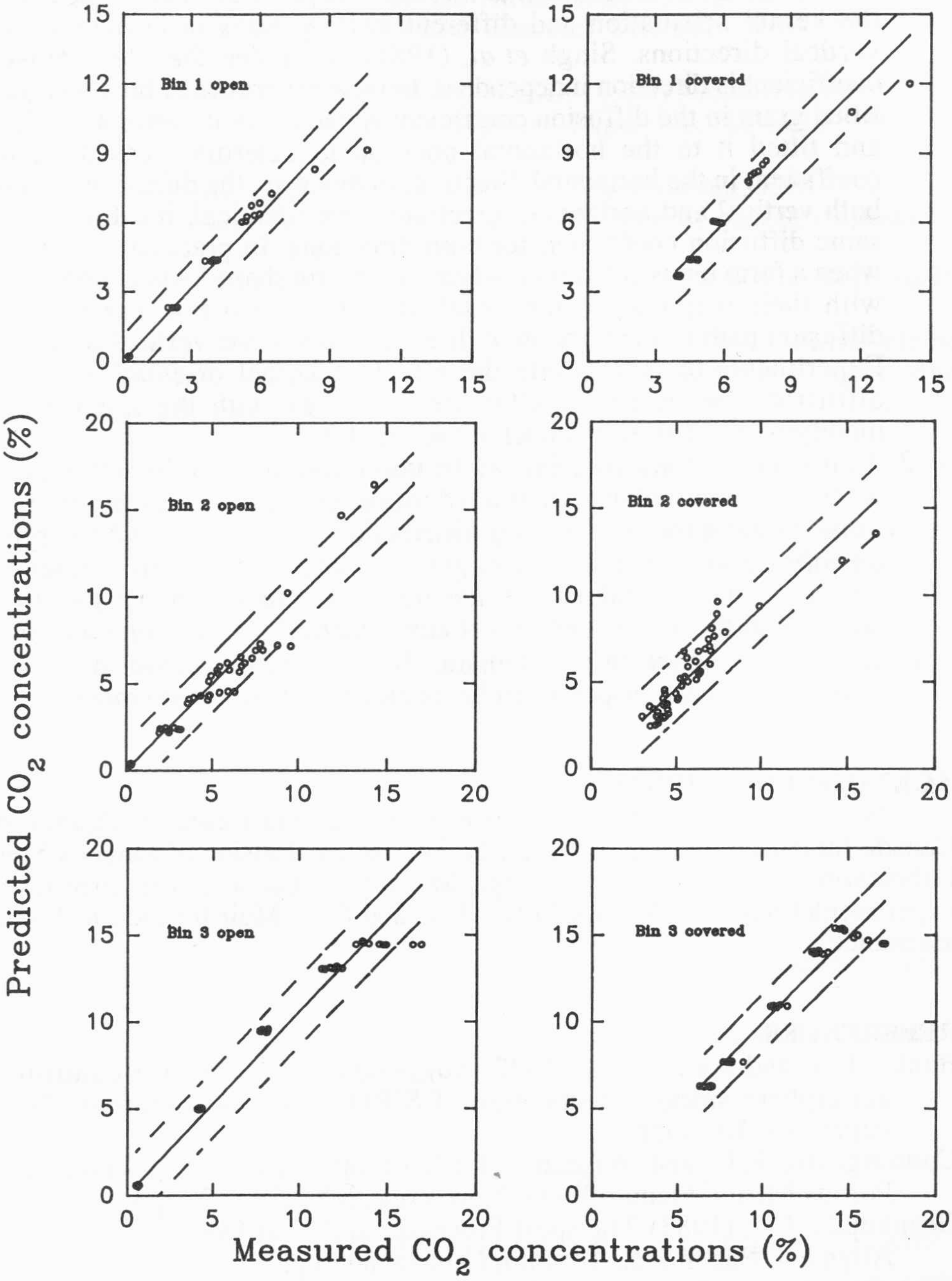

Fig. 3: Measured and predicted CO2 concentrations at 21 h. The dashed lines show the 95% confidence intervals for the individual predicted values.

shown that the airflow resistance of grains is higher in the vertical direction than in the horizontal direction (Jayas *et al*., 1987) because of the kernel orientation and different airflow paths in horizontal and vertical directions. Singh *et al*. (1984) concluded that the diffusion coefficient is direction independent. In their experiments, however, they filled grain in the diffusion coefficient apparatus in its vertical position and tilted it to the horizontal position to determine the diffusion coefficient in the horizontal direction. In this way, the diffusion path for both vertical and horizontal directions were identical, resulting in the same diffusion coefficient for both directions. In practical situations, when a farm bin is filled with wheat, the oblate shaped wheat kernels lie with their major axes horizontal, thereby creating a less resistant diffusion path in the horizontal direction than in the vertical direction. Experiments to incorporate the effect of kernel orientation on the diffusion coefficient of CO_2 are underway with the intention of modifying the diffusion model in the near future.

2. To use the 1 h measured data as the initial condition in the simulations, we linearly interpolated the limited measured data to every point in the finite element mesh. The CO_2 distribution at 1 h may not be varying linearly between nodes, and might have caused errors in prediction, especially in the initial periods. Ideally, the simulation should start from time 0 with the initial condition of atmospheric CO_2 concentration (0.55 g/m^3) everywhere in the domain. To do this, the model of forced convective mass transport must be included in the diffusion model.

ACKNOWLEDGEMENTS

We thank the Natural Sciences and Engineering Research Council of Canada for partial funding of this study; Westeel, a Division of Jannock Steel Fabricating Company, Winnipeg, Manitoba, Canada for providing experimental bins; and Messrs. D.D. Mann and D. P. Muir for their technical assistance.

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LIST OF SYMBOLS

A & B	Empirical constants
c -	CO_2 concentration, kg/m^3
cc -	CO_2 concentration in the box, % (in Eq. 6)
C_i -	initial CO_2 concentration in the grain bulk, kg/m^3
Cs_1, Cs_2 -	CO_2 concentrations specified on the boundaries S_1 and S_2, respectively, kg/m^3
D_x, D_y, D_z -	diffusion coefficient of CO_2 through grain in x, y and z coordinate directions, respectively, m^2/s
e -	mean relative percent error of prediction, % (in Eq. 7)
ℓ -	direction cosines
M_i -	measured CO_2 concentration at sampling point i, kg/m^3
n -	number of data points
P_i -	predicted CO_2 concentration at sampling point i, kg/m^3
q -	amount of CO_2 adsorbed or produced by the grain, kg/m^3
Q -	surface flux across the boundary S_3, kg/(m^2.s)
t -	time, h (in Eq. 6)

Ω - problem domain

τ- time, s (in Eq.1)